REMARKS

I.   Status of Claims

Claim 1 is amended to incorporate the subject matter of claim 2, and to define that the

two peaks in the particle size distribution correspond to two different particle sizes.

Claim 2 is canceled without prejudice or disclaimer.

No new matter is added. Accordingly, Applicants respectfully request entry and

consideration of the Amendment. Upon entry of the Amendment, claims 1 and 5-14 will be

pending.

II.   Response to Claim Rejections Under 35 U.S.C. § 103(a)

A.   Claims 1, 2, 8, 9, 12 and 13 are rejected under 35 U.S.C. § 103(a) as allegedly

being unpatentable over Hwang et al. (WO 99/37592; "Hwang I") in view of Goodwin (U.S.

Patent No. 3,331,671) and Kulkarni (Studies on Fly Ash-Filled Epoxy-Cast Slabs Under

Compression, J. Appl. Polymer Science, 84, 2404-2410 (2002)).

B.   Claims 5-7 are rejected under 35 U.S.C. § 103(a) as allegedly being unpatentable

over Hwang I in view of Goodwin, Kulkarni, Chang (U.S. Patent No. 5,505,766) and Nomura et

al. (JP 2001-220193; machine translation).

C.   Claims 9, 10 and 14 are rejected under 35 U.S.C. § 103(a) as allegedly being

unpatentable over Hwang I in view of Goodwin, Kulkarni and Hwang (U.S. Patent No.

5,047,145; "Hwang II").

D.   Claim 11 is rejected under 35 U.S.C. § 103(a) as allegedly being unpatentable over Hwang I in view of Goodwin, Kulkarni, Chang and Nodera et al. (U.S. Patent No. 5,837,757).

Applicants respectfully traverse, at least for the following reasons.

Present claim 1 recites a flame-retardant resin composition comprising a polycarbonate type resin and fly ash which contains particles composed of a complex of silicon dioxide and aluminum oxide and has a 50% particle size (D50) of 1 to 10 μm and has two peaks in its particle size distribution, wherein the peak corresponding to the particle size distribution of larger particles is greater than the peak corresponding to the particle size distribution of smaller particles. The amount of fly ash in the total composition is from 1 to 60 weight %. The content of 1 to 60 weight % of fly ash achieves suitably flame retardancy and mechanical strength of polycarbonate composition in this application.

The rejection of claim 1, and dependent claims thereof, based on Hwang I was addressed in the Amendment under 37 C.F.R. § 1.116, filed June 17, 2009. None of Goodwin, Chang, Nomura and Nodera cured the deficiency in Hwang I, with respect to the bimodal particle size distribution, as presently claimed.

The Examiner relies on Kulkarni to teach a fly ash composition in which there is a bimodal distribution where the peak corresponding to the larger particles is greater than the peak corresponding to the smaller particles.

In Hwang I the fly ash and calcium carbonate have the same mechanical properties as filler in many plastics. However, with regard to flame retardancy against polycarbonate they do

not behave in a similar manner. See, for example, M. Soyama et al. (Polymers for Advanced Technologies, Vol. 18, 386-391(2007)), a copy of which accompanies the Amendment. Calcium carbonate has no flame retardancy, as discussed in the Results and Discussion section of Soyama. See, for example, the first full paragraph on page 388, which states "calcium carbonate... did not make the polycarbonate more flame retardant."

Kulkarni uses a bimodal fly ash having a mean size of about 21 μm, as shown in Fig. 2, where the peak corresponding to the larger size particles is at 35 μm and 73% and the peak corresponding to smaller size particles is at 0.35 μm and 27%. The particle sizes of Kulkarni are therefore, outside the D50 particle size range of present claim 1, i.e., 1 to 10 μm. In the Conclusion section, Kulkarni describes that high filling is achieved by the use of the bimodal fly ash in epoxy resin with a maximum packing volume of 0.36. However, Kulkarni does not teach that for achieving flame retardancy and mechanical strength of the polycarbonate resin, fly ash having a D50 particle size of 1 to 10 μm should be added to the polycarbonate resin.

Therefore, claim 1 is patentable over any combination of Hwang I, Goodwin, Kulkarni, Chang, Hwang II, Nomura and Nodera. Claims 5-14 are also patentable, at least by virtue of their dependence from claim 1. Accordingly, Applicants respectfully request reconsideration and withdrawal of the § 103(a) rejections of claims 1 and 5-14.

Conclusion

In view of the above, reconsideration and allowance of this application are now believed to be in order, and such actions are hereby solicited. If any points remain in issue which the
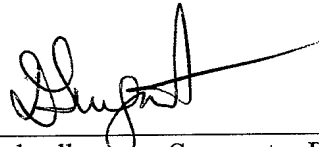
Examiner feels may be best resolved through a personal or telephone interview, the Examiner is

kindly requested to contact the undersigned at the telephone number listed below.

The USPTO is directed and authorized to charge all required fees, except for the Issue

Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any

overpayments to said Deposit Account.

Respectfully submitted,

Debodhonyaa Sengupta, Ph.D.
Limited Recognition No. L0578

SUGHRUE MION, PLLC
Telephone: (202) 293-7060
Facsimile: (202) 293-7860

WASHINGTON OFFICE
23373
CUSTOMER NUMBER

Date: June 3, 2010

PAT

Flame retardancy of polycarbonate enhanced by adding fly ash

Makoto Soyama*, Kazuhiko Inoue and Masatoshi Iji

Fundamental and Environmental Research Laboratories, NEC Corporation, 34 Miyukigaoka, Tsukuba, Ibaraki 305-8501, Japan

We have developed a flame-retardant polycarbonate resin whose flame retardancy is greatly enhanced by adding fly ash (a by-product from thermal power plants). Fly ash with comparatively small average particle sizes (<10 μm) raised the flame retardancy of polycarbonate because of the hydrogen bond that forms between the polycarbonate and the hydroxy group on the fly ash surface. This bond primarily improves the heat resistance of the polycarbonate and also isomerizes the polycarbonate to promote carbonization. The polycarbonate with 25 wt% of the fly ash showed good other main characteristics, such as mechanical properties and moldability, comparable to those of a flame retardant polycarbonate reinforced with glass fiber, used for electronic products such as desktop computers, etc. Applying the fly ash eliminates the need to use current retardants such as organic halogen compounds and thus promotes the perceived environmental safety of flame-retardant polycarbonate. In addition, being able to use less raw resin by adding the fly ash means that less energy is required to manufacture flame-retardant polycarbonate. Copyright © 2007 John Wiley & Sons, Ltd.

KEYWORDS: polycarbonates; fly ash; flame retardance; degradation; inorganic materials

INTRODUCTION

Flame retardancy is an essential property for preventing the plastics used in the housing of electronic products, such as polycarbonate and polystyrene, from accidental burning. Hence, organic halogen compounds have been added to these plastics as flame retardants. Incinerating plastics containing such as organic halogen compounds, however, generates halogen gases, creating a major environmental safety hazard. Therefore, phosphorus compounds, sulfonates, and silicone compounds, which are safer but less effective as flame-retardants than the halogen compounds, are now being used as flame retardants for polycarbonate, which has comparatively high flame redundancy compared with other plastics such as polystyrene and its derivatives used in housing.[1] The manufacture of polycarbonate, however, consumes more energy and is more expensive than the manufacture of other applicable plastics. The energy required and the cost must be reduced to expand the use of polycarbonate.

Combining plastics and inorganic fillers requiring a little energy to produce them is an effective way to reduce the energy required for manufacturing the plastics. The inorganic compounds typically used as fillers (such as silica, calcium carbonate, and talc) also make plastics slightly less flammable.[2-7] To significantly increase the flame retardancy

of plastics, though, large amounts of these fillers must be added and such large amounts lower the fluidity and strength of the plastics. This makes such plastics unsuitable for the housing of electronic equipment. In addition, the overall energy consumption is not greatly reduced since energy is used in the manufacture of the inorganic compounds.

In response to this problem, we have examined the use of fly ash: a by-product from thermal power plants as a new flame retardant for polycarbonate. Fly ash, composed of mainly silica and alumina, is a by-product obtained when ground coal is fed into burners to generate power and shows dark color because of unburned carbon content. As a result, little energy is needed to produce it, and in Japan this corresponds to designating a by-product as a renewable resource. A huge amount of fly ash is generated worldwide; in Japan alone, the annual production was about seven million tons in 2000.[8]

Some fly ash is used in the construction industry, mainly in cement and concrete manufacturing, while the remainder is disposed of in landfill sites. Using fly ash in a beneficial way would be preferable to dumping it, so many researchers have been looking for useful applications. One example is to mix fly ash with plastics to lower production costs; this has been done in the commercial production of commodity plastics such as polypropylenes.[9] This research has shown that

*Correspondence to: M. Soyama, Fundamental and Environmental Research Laboratories, NEC Corporation, 34 Miyukigaoka, Tsukuba, Ibaraki 305-8501, Japan.
E-mail: m-soyama@cj.jp.nec.com

WILEY
InterScience®
DISCOVER SOMETHING GREAT

adding fly ash to plastics leads to decreased moldability as well as lower manufacturing costs, but the particular characteristics of fly ash as a flame retardant have not been much examined.

Thus, we have investigated the flame- retarding characteristic of polycarbonate with added fly ash. In this paper, we describe the flame- retarding characteristic and its underlying mechanism as well as other main characteristics such as mechanical properties and moldability of this resin compound.

EXPERIMENTAL

Materials

As a thermoplastic, we used polycarbonate (Sumitomo Dow Co., Ltd, Caliber 301-22). The fly ash (Yonden Business Industry Co., Yonden fly ash and Fynash FA20) that we used had the consistency of a very fine powder with primarily spherical particles with average particle sizes of about 5 and 20 μm (Fig. 1). The fly ash primarily consisted of inorganic materials such as silica, alumina, and calcium oxide. Table 1 shows the chemical characterization of typical fly ash. Three other inorganic materials were used in this study (Table 2):
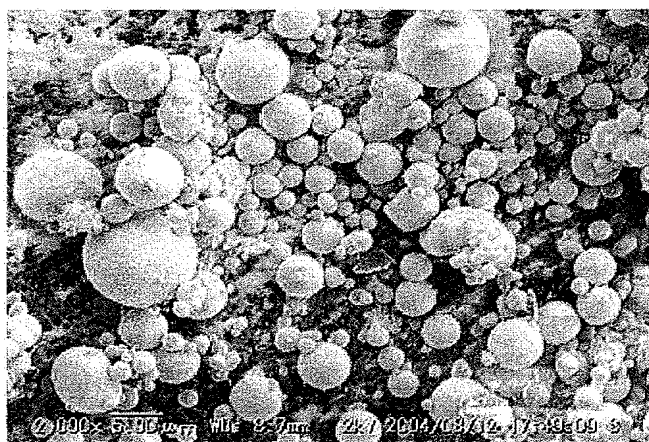

Figure 1. Scanning electron microscopy image of fly ash.

Table 1. Chemical characterization of typical fly ash

Source	Unit	Content
SiO_2	wt%	67.89
Al_2O_3	wt%	21.92
Na_2O	wt%	0.36
K_2O	wt%	1.36
SO_3	wt%	0.42
Fe_2O_3	wt%	3.94
CaO	wt%	0.57
MgO	wt%	0.63
MnO	wt%	0.03
TiO_2	wt%	1.28
igross	wt%	1.3
pH	—	7.4
Specific surface	m^2/g	3.5
Density	g/cm^3	2.21

spherical silica (Denki Kagaku Kogyo K. K., FB 5604, FB 24, FB 3 SDC, SFP 30 M), spherical alumina (Admatechs Co., Ltd, AO-509 and Showa Denko K. K., CB-A05S), and spherical calcium carbonate (Komesho Sekkai Kogyo Co., Ltd, ED-SIII). We used trioctyl trimellitate (Daihachi Chemical Industry Co., Ltd, TOTM) as a plasticizer. We also added 0.5 wt% of polytetrafluoroethylene (Daicel Chemical Industries, Ltd, Polyflon MPA FA 500) to prevent dripping during heating.

Methods

Sample compounds were prepared by mechanically kneading the polycarbonate with inorganic fillers and the other additives using an extruder (KCK Co., Ltd, KCK80 × 2-35VE × 5) at 280°C. Test pieces of the compounds for evaluating the flame retardancy and the flexural property were molded with an injection molding machine (Toshiba Machine Co., Ltd, EC20P-0.4A), at a cylinder temperature of 280°C and a mold temperature of 80°C.

We measured the flame retardancy of the molded compounds by the oxygen index method (JIS-K-7201) and the UL94V method of Underwriters Laboratories (UL), Inc. The oxygen index indicates the oxygen concentration required for the material to continue burning. In the UL94V method, a vertically clamped sample of the molded compound is ignited by a gas burner flame applied twice for 10 sec to the bottom of the sample. For a material to be classified as V-0 requires that burning stops within 10 sec after two ignitions. The V-1 and V-2 classifications require that burning stops within 60 sec after two ignitions, but V-2 allows a flaming drip. In all tests, the samples cannot be allowed to burn out.

We measured the flexural strength of the compounds and their modulus with a versatile examination machine (Instron Co., Ltd, 5567), under the conditions of a 50-mm distance between the supporting points and a 1.6-mm/min crosshead speed. Their Izod impact strength with a notch in the resin was measured with a universal impact tester (Orient Precision Machine Ltd, C1).

The moldability of the compounds was evaluated according to the melt flow characteristic, which was evaluated along the flow length (spiral flow) of the compounds in a metal molding. The plasticization conditions were set so that an injection temperature of 280°C was achieved. The mold temperature was maintained at 80°C. The injection rate was 100 mm/sec, and the thickness of the product was 1 mm.

The thermal stability of the compounds was measured in air (200 ml/min) at a heating rate of 10°C/min by thermogravimetric analysis (EXTAR6000 TG/DTA, Seiko Instruments Co.).

The pyrolysis gas of the compound (0.5–1 mg) was identified by mass spectrometry. In addition, we calculated the amount of generated gas as compared with the peak area of a GC-MASS (QP-5050A [GC-17A], Shimadzu Co.) chart. The heating temperature was set to 800°C for the thermal decomposition division, a helium gas atmosphere was used in the heating treatment, and the interface temperature was measured at 300°C.

Table 2. Characteristics of the inorganic compounds used in this study

Inorganic compound	Source	Content (%)	Specific surface (m^2/g)	Average particle diameter (μm)
Fly ash	SiO_2	Table 1	3.5	20
	Al_2O_3		5.5	5
Spherical silica	SiO_2	100	2	32
			2.5	18
			2.4	11
			3.7	3
			5	0.6
Spherical alumina	Al_2O_3	>99.8	0.6	10
			1.4	2.8
Spherical calcium carbonate	$CaCO_3$	93.0	7	3

RESULTS AND DISCUSSION

Flame-retarding effect of inorganic filler for polycarbonate

We compared the effects of the kind and particle size of the inorganic fillers on the flame retardancy of polycarbonate (Fig. 2). The results confirmed that silica, alumina, and fly ash, which all have hydroxy groups on their surface, made the polycarbonate more flame retardant. Fly ash with small particle sizes and wide specific surfaces tended to make the polycarbonate highly flame retardant. In particular, fly ash with an average particle size of 10 μm or less greatly improved the flame retardancy of the polycarbonate compared to the effect of the other fillers. On the other hand, calcium carbonate, with few hydroxy groups on its surface, did not make the polycarbonate more flame retardant.[10] In addition, increasing the quantity of fly ash with an average particle size of about 5 μm improved the flame retardancy of the polycarbonate (Fig. 3).

Flame-retarding mechanism for polycarbonate with fly ash

We clarified the flame-retarding mechanism for polycarbonate with fly ash by evaluating the progress of carbonization, through quantitative analysis of thermal decomposition products, and from the change in the thermal degradation. We also conducted a similar evaluation using added silica as a reference, because silica is the main component of the fly ash.

Progress of carbonization
We first studied how the carbonization of polycarbonate is promoted by adding fly ash. Ballistreri *et al.* show the change

in the chemical structure of the polycarbonate as it carbonizes; through this change, the proportion of carbon increases relative to that of hydrogen.[11] We evaluated the progress of the carbonization by observing the material around the ignition area and measuring the fixed quantities of carbon and hydrogen. We found that the fly ash promoted carbonization of the polycarbonate, since both the fly ash and the silica reference increased the ratio of carbon to hydrogen in the compounds containing them after ignition (Table 3).

Quantitative analysis of thermal decomposition products
The thermal decomposition products of the polycarbonate with added fly ash or silica were quantitatively analyzed. Table 4 lists the thermal decomposition gases, and Fig. 4 shows the results of gas chromatography mass spectrometric analysis (GC-MASS). The gas components and generation rates showed that the total quantity of thermal decomposition gas decreased by about 60% because of the presence of fly ash or silica, which reduced the generation of phenols and bisphenol A in the middle-molecular-weight portion in the thermal decompositions of the polycarbonate (Table 5). This suggests that fly ash or silica changes the thermal decomposition mechanism of the polycarbonate, and that the decomposition products do not gasify but rather are fixed as a char through the dehydration condensation reaction, indicating that the polycarbonate is made more flame retardancy.

We deduced the fragmentation mechanism of the polycarbonate with added fly ash from the thermal decomposition analysis. The polycarbonate decomposes thermally through reduction of the isopropylidene group or the
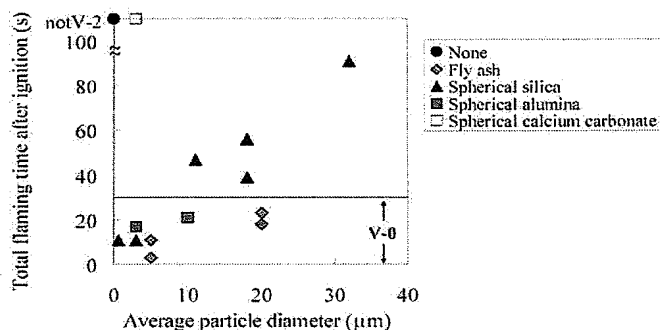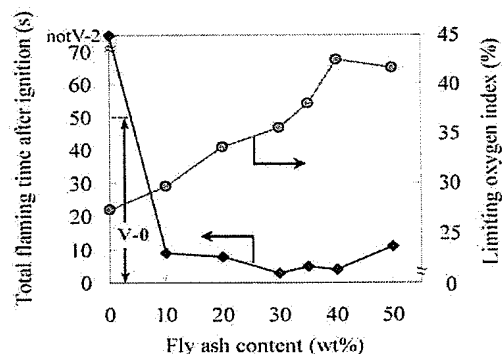

Figure 2. Flame-retarding effect of polycarbonate with 30 wt% of inorganic compounds.

Figure 3. Flame-retarding effect of fly ash (particle size 5 μm) added to polycarbonate.

Table 3. Changes in the C/H ratio caused by ignition for polycarbonate with fly ash

Sample	Ignition	C/atm%	H/atm%	C/H
Polycarbonate	Before	48.0	43.1	1.11
Polycarbonate	After	48.6	42.6	1.14
Polycarbonate with 30 wt% fly ash	After	49.1	40.1	1.22
Polycarbonate with 30 wt% spherical silica	After	48.3	37.1	1.30

carbonate group, as proposed by some authors.[12–16] The division mechanism shows how the generation of phenols reduces the carbonate group. The fact that fly ash or silica reduced the phenols in the thermal decomposition gas of the polycarbonate shows that these additives inhibited reduction of the carbonate group and promoted reduction of the isopropylidene group.

Thermal degradation

We used thermogravimetric analysis to investigate the effect of fly ash on the thermal degradability of the polycarbonate (Fig. 5). The results show that the fly ash improves resistance to the degradability. This reason for this appears to be that the fly ash stabilizes the carbonate group of the polycarbonate through the strong hydrogen bond between the hydroxyl group on the fly ash surface and the carbonate group, and then retards pyrolysis of the polycarbonate.

Proposed flame-retarding mechanism of fly ash

Based on the above results, here we propose a mechanism to explain the flame-retarding effect that fly ash has on polycarbonate. Of the inorganic fillers we tested, only those including fly ash and silica, which have hydroxy groups on their surfaces, made polycarbonate more flame retardant. We also showed that the fly ash increased the thermal degradation resistance of polycarbonate. Furthermore, because fly ash and silica reduced the middle-molecular-weight component in the thermal decomposition of the polycarbonate, they accelerated the dehydration condensation between the phenols and the bisphenol A to promote the carbonization of the polycarbonate.[17–18] Therefore, we believe the hydrogen bond between the hydroxy group on the fly ash surface and the carbonate group of the polycarbonate is the cause of the flame retardancy of the polycarbonate (Fig. 6). That is, this bond thermally stabilizes the polycarbonate on the fly ash surface, thus improving heat

resistance; it also isomerizes the polycarbonate, promoting carbonization through which the fly ash improves the polycarbonate flame retardancy.

We thus concluded that the flame-retarding mechanism of the polycarbonate added fly ash consists mainly of the two following steps:

I. The hydrogen bond between the hydroxy group on the fly ash surface and the carbonate group of the polycarbonate stabilizes the combination in the polycarbonate, thus
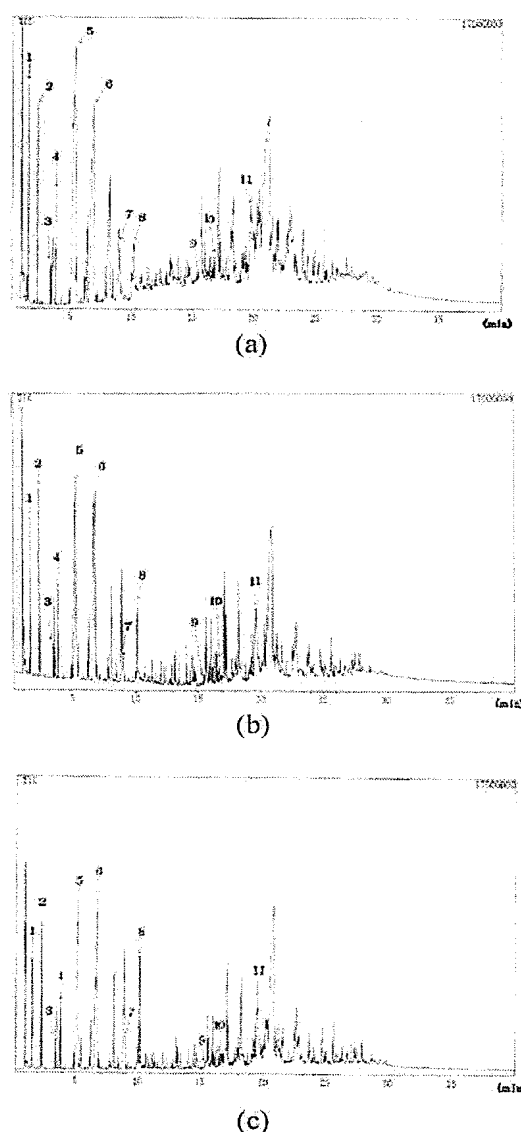
(a)

(b)

Table 4. Thermal decomposition products assigned by mass spectroscopy

Peak No.	Structure
1	Benzene
2	Toluene
3	Phenol
4	p-Cresol
5	o-Cresol
6	p-Ethyl phenol
7	Isopropyl phenol
8	Ally phenol
9	Diphenyl carbonate
10	Carbonize acid 4-methyl ester
11	4,4'-Methylenebis-phenol

(c)

Figure 4. GC-MASS charts of (a) polycarbonate, (b) polycarbonate with 30 wt% fly ash, and (c) polycarbonate with 30 wt% spherical silica.

Table 5. Comparison of the amounts of thermal decomposition products measured by GC-MASS (800°C)

	Benzene	Toluene	Benzene derivatives	Phenol	Cresol	Isopropyl phenol	Phenolic derivatives	Bis phenolic derivatives	Gas generation
Polycarbonate	100	100	100	100	100	100	100	100	100
Polycarbonate with 30% of fly ash	105	124	115	73	129	53	85	48	59
Polycarbonate with 30% of spherical silica	94	128	111	72	124	27	74	54	67
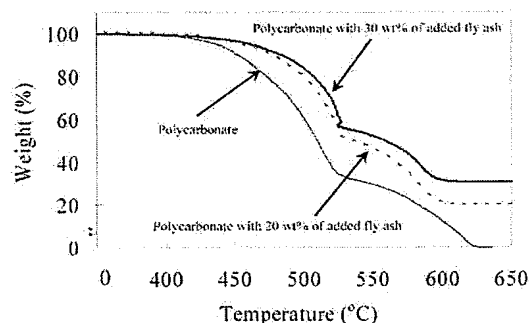
**Figure 5.** Thermogravimetric analysis of polycarbonate containing fly ash (air: 200 ml/min, 10°C/min).

improving its heat resistance, while also isomerizing the polycarbonate.

II. The isomerized polycarbonate is condensed during dehydration with the phenols in the decomposition product, becoming carbonized to form a material like char, which is advantageous for flame retardation.

Figure 3 shows that the polycarbonate flame retardancy is more improved by fly ash than by the silica and alumina, which also have the hydroxyl group. This effect can be expected even if the flame retardancy is actually improved by a different factor specific to the fly ash; for example, if the silica-alumina composite included in the fly ash showed higher acidity than plain silica and alumina, thus increasing its effect as a solid acid catalyst to promote charring of the polycarbonate.[19]

Mechanical characteristics and moldability of polycarbonate with fly ash

Figure 7 shows the flexural properties of polycarbonate with fly ash, and Fig. 8 shows its impact strength and fluidity. The flexural modulus improved with increasing fly ash content.
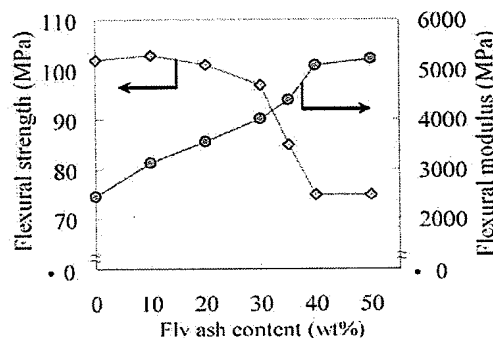
Figure 7. Flexural properties of polycarbonate containing fly ash.

While the flexural strength was maintained at comparatively low contents of fly ash, it fell sharply when the content exceeded 30 wt% (Fig. 7). Adding too much fly ash to the polycarbonate also lowered its impact strength. These characteristics of the polycarbonate with 25 wt% of added fly ash were almost equal, however, to those of a
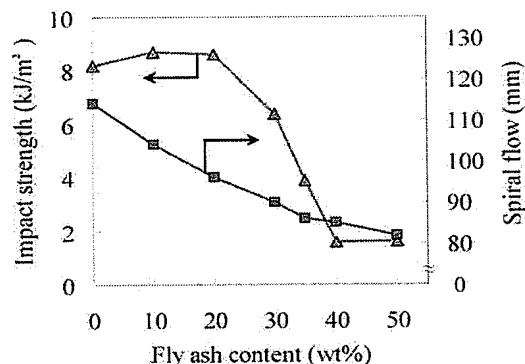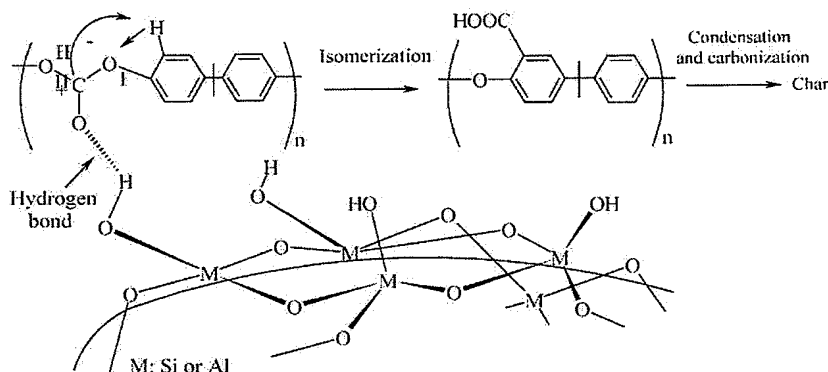

Figure 8. Impact strength and fluidity of polycarbonate containing fly ash.

Figure 6. Influence of hydrogen bonding on isomerization and carbonization.

Table 6. Properties of polycarbonate compounds containing fly ash

	Unit	Polycarbonate	PC with 10% glass fiber*	PC with 25% fly ash	PC with 25% fly ash and 5% plasticizer
Flame retardant (UL-94. 1/16 in.)	—	V-2	V-0	V-0	V-0
HDT ($^{\circ}$C)	$^{\circ}$C	134	132	128	98
Spiral flow (280°C, 1 mm-t)	mm	120	110	100	157
Flexural strength	MPa	90	98	99	108
Flexural modulus	MPa	2250	2940	3401	3862
Impact strength	kJ/m^2	75	8	7	5

*Flame-retardant polycarbonate with a silicone flame retardant and glass fiber (10%).

polycarbonate with 10 wt% glass fiber, which is currently used as a housing material for electronic products such as desktop computer, etc.

Though fluidity is lowered with this quantity of fly ash added to the polycarbonate, this was improved by adding a plasticizer with comparatively high heat resistance, such as trioctyl trimellitate, while maintaining its flame retardancy, as shown in Table 6.

The appearance of the polycarbonate with fly ash was somewhat dark color owing to the original color of fly ash, which can retard the use in products required to be clear colors.

CONCLUSION

We found that fly ash, a by-product of thermal power plants, greatly enhances the flame retardancy of polycarbonate and thus decreases the amount of energy required to manufacture flame-retardant polycarbonate. The flame retardancy of the polycarbonate with fly ash results from the hydrogen bond between the hydroxy group on the fly ash surface and the carbonate group of the polycarbonate. This bond thermally stabilizes the polycarbonate on the fly ash surface and thus improves its heat resistance. In addition, it isomerizes the polycarbonate and then promotes carbonization, which improves flame retardancy. Though adding a large amount of fly ash degrades the mechanical characteristics and moldability of the material, the characteristics of polycarbonate with 25 wt% of fly ash were equal to those of polycarbonate with 10 wt% glass fiber, which is used as a housing material for electronic products such as desktop computers, etc. In addition, combining this material with a plasticizer having high heat resistance drastically improves its fluidity, while maintaining its flame retardancy.

REFERENCES

1. Iji M, Serizawa S. Silicone derivatives as new flame retardants for aromatic thermoplastics used in electronic devices. *Polym. Advan. Technol.* 1998; **9**: 593.
2. Gilman JW, Kashiwagi T. *Flame retardant mechanism of silica. 9th Int. Conf. Addit.* 2000; 55.
3. Kashiwagi T, Shields JR. Flame-retardant mechanism of silica: effects of resin molecular weight. *J. Appl. Polym. Sci.* 2003; **87**: 1541.
4. Chand N, Vashishtha SR. Development, structure and strength properties of PP/PMMA/FA blends. *B. Mater. Sci.* 2000; **23**: 103.
5. Nikkesi S, Ishiyama H. The effects of addition of surface-treated silica particle in polycarbonate composites. *J. Jpn. Soc. Polym. Process.* 1998; **10**: 891.
6. Kashiwagi T, Gilman JW. Flame retardant mechanism of silica gel/silica. *Fire Mater.* 2000; **24**: 277.
7. Levchik GF, Levchik SV. Mechanisms of action in flame retardant reinforced nylon 6. *Polym. Degrad. Stabil.* 1996; **54**: 361.
8. Nagataki S, Tomosaka F, Kanazu T, Yamamoto T. Classification of fly ash according to japan industrial standard, JIS 6201-1999. *Spec. Publ. Am. Concr. Inst.* 2001; **1**: 29.
9. Kondoh M, Mori S. Effective utilization of ash from fluidized-bed by adding it to polypropylene resin. *J. Chem. Eng. Jpn.* 1999; **25**: 374.
10. Fuji M. Functionalization of particle surface. *Sur. Sci. Soc. Jpn.* 2003; **24**: 625.
11. Ballistreri A, Montaudo G. Intumescent flame retardants for polymers. IV. The polycarbonate-aromatic sulfonates system. *J. Polym. Sci. Part A1.* 1988; **26**: 2113.
12. Ishikawa T, Maki I. Effect of perfluoroalkane sulfonic acid on the flame retardancy of polycarbonate. *J. Macromol. Sci. Pure.* 2004; **41**: 523.
13. McNeil IC, Rincon A. Thermal degradation of polycarbonates: reaction conditions and reaction mechanisms. *Polym. Degrad. Stabil.* 1993; **39**: 13.
14. Montaudo G, Puglisii C. Thermal decomposition processes in bisphenol a polycarbonate. *Polym. Degrad. Stabil.* 1992; **37**: 91.
15. Marks MJ, Erskine JS. Products and mechanism of the thermal cross-linking of benzocyclobutene-terminated bisphenol a polycarbonates. *Macromolecules* 1994; **27**: 4114.
16. Ito Y, Ogasawara H. Characterization of end groups in polycarbonates by reactive pyrolysis-gas chromatography. *Polym. J.* 1996; **28**: 1090.
17. Tesoro GC. Fire resistance in advanced engineering thermoplastics. *ACS. Sym. Ser.* 1990; **425**: 241.
18. Davies A, Golden JH. Thermally-induced cross-linking of poly[2.2-propane-bis-(4-phenyl carbonate)]. *Macromol. Chem.* 1967; **110**: 180.
19. Shiba T, Sato M, Hattori H, Yoshida K. Acidities of silica-alumina catalysts. *Catalysts Catal.* 1964; **6**: 80.